

Supplemental Material: 'Linear and nonlinear rheology of dense emulsions across the glass and the jamming regimes'.

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Yield stress behavior.

Here we provide evidence that the shear stress measured for purely repulsive emulsions does not show history dependence even in a regime around and below the jamming transition ϕ_J , where thermal effects play an important role. In Figure S1 we show recent rheology experiments for silicone oil in water emulsions with a droplet size $R=430\text{nm}$, $\approx 10\%$ polydispersity, stabilized by 10mM SDS and with 10 mM NaCl added (Debye length $\lambda_D \approx 2.3\text{ nm}$). This system behaves very similarly to the emulsion discussed in the main text and shows a jamming transition $\phi_J^{eff} \sim 0.64$. Starting from a highly sheared state, we reduce the shear rate as shown in Figure S1. The shear stress of the emulsion at $\phi_{eff} \sim 0.63$ decreases (black circles). Subsequently, we increase the shear rate, and the measured shear stress remains on the same curve (red triangles). Next, we decrease the shear rate, and the shear stress again follows the same curve (green squares). This measurement clearly demonstrates that the emulsion is a “simple” yield stress fluid¹, since no indication for hysteresis or history dependence is found. We thus conclude that the yield stress is well defined for our system.

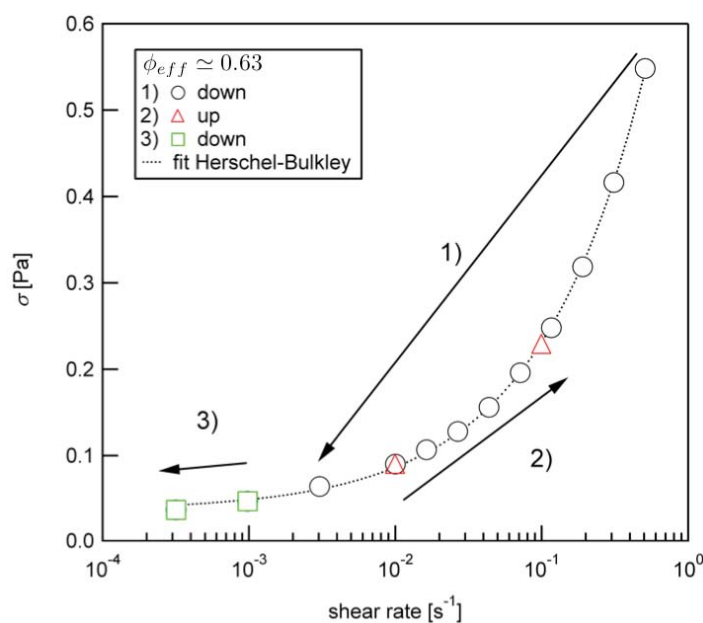


Figure S1 Flow curve for an emulsion with $\phi_{eff} \sim 0.63$ obtained by increasing and decreasing the shear stress. The dotted line shows an empirical Herschel- Bulkley fit $\sigma = \sigma_y + K\dot{\gamma}^\beta$ to the data with $\sigma_y = 0.033\text{Pa}$ ($K = 0.77$ and $\beta = 0.59$ are adjustable parameters and $\dot{\gamma}$ is the shear rate).

¹ P. Moller, A. Fal, V. Chikkadi, D. Derks and Daniel Bonn, *An attempt to categorize yield stress fluid behaviour*, Phil. Trans. R. Soc. A **367**, 5139-5155 (2009)

Comparison to a model with an interaction potential of the form $V(r) = \epsilon(1 - r/2R)^\alpha$

The simplest way to model the jamming of dense assemblies of soft spheres is to assume pairwise interactions of the type $V(r) = \epsilon(1 - r/2R)^\alpha$, e.g. with $\alpha=2$ (harmonic) and $\alpha=2.5$ (Hertzian). Numerical studies typically are based on such an interaction potential. Our equation (5), $V(r) = 3^{-\alpha}\epsilon[(2R/r)^3 - 1]^\alpha$, has been determined by Lacasse, Grest and co-workers² as an effective interaction potential for actual emulsion droplets with $\alpha \approx 2.32$. Eq(5) reduces to the simpler form (commonly employed in simulations) close to ϕ_j but deviates at larger densities. In Figure S2 we compare the predictions of the simpler potential leading to $k(\phi) \sim (\phi - \phi_j)^{\alpha-2}$ with our predictions based on the full Eq.(5) and (6). The plot reveals that the agreement is fairly good for $\phi \leq 0.7$ (for any of the exponents) but poor for densities above $\phi \geq 0.7$. Using an adjustable prefactor and varying ϕ_j^{eff} does not result in a substantial improvement of the fit. For a quantitative description we have thus had to use the full expression $V(r) = 3^{-\alpha}\epsilon[(2R/r)^3 - 1]^\alpha$.

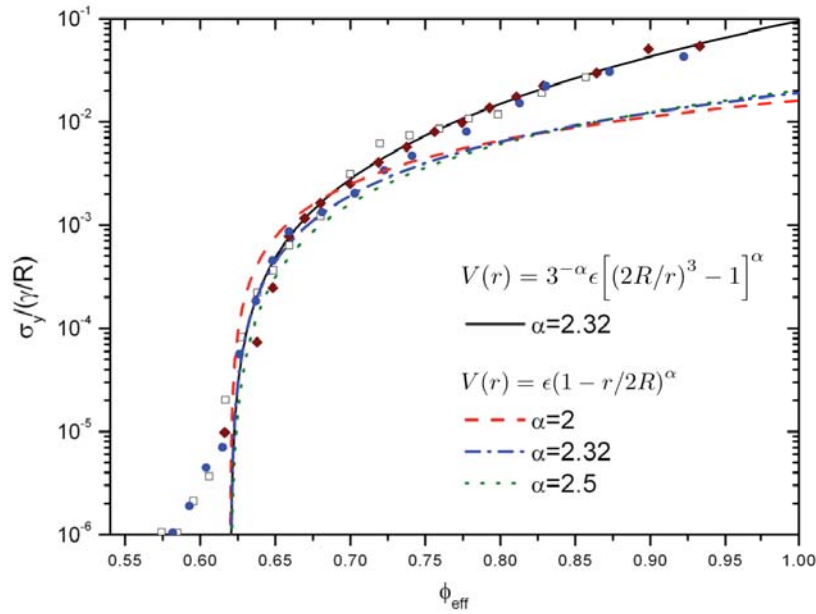


Figure S2 Yield stress of emulsions as reported in Fig 3. Solid line shows the yield stress as discussed in the main text (based on Eq.(5)). Dashed, Dash-dotted and dotted lines show the predictions using the simpler expression for the interaction potential using exactly the same parameters.

Scaling relation for the yield stress

There appears to be a broad consensus concerning the power law scaling of the shear modulus with an exponent 0.5 for harmonic spheres (see. e.g. our references [1,2]). Predictions about scaling of the yield stress however are more recent and the results are still under discussion. The numerical results by Olsson & Teitel cited in the main text suggest an exponent 1.2 for harmonic spheres. The “Granular Rheology” paper by Hanato (J. Phys. Soc. Jpn **77**, 123002, 2008) equally suggests an exponent 1.2 (their Figure 2 a) for harmonic spheres. Otsuki and Hayakawa suggest an exponent of 1, although the

² M-D. Lacasse, G. S. Grest, D. Levine, T. G. Mason, and D. A. Weitz, *Phy. Rev. Lett.* 76, 3448 (1996)

data shown in their Figure 5 (Phys. Rev. E **80**, 011308, 2009) is very limited, based on only two data points close to Φ_J . The article by Tighe et al. (PRL **105**, 175701 (2010)) suggests a scaling with an exponent of 1.5 for the yields stress (harmonic spheres, their Figure 3).

In Figure S3 we compare our model predictions and the experimental data for exponents $\delta = 1, 1.2$ and 1.5 , $\sigma_y(\phi) \sim k(\phi)(\phi - \phi_J)^\delta$. We do not alter the model prediction for $G_p(\phi)$ given by Eq. (2) and (7) and thus with Eq.(3): $\sigma_y(\phi) = (a_2/a_1)G_p(\phi)(\phi - \phi_J)^{\delta-0.5}$. Adjusting the parameter a_2 allows a decent fit to the data for all these exponents. Adjusting δ and a_2 however adversely affects the predictions for the yield strain, Figure 1 ($a_1 \cong 0.2$ is kept constant). As show in Figure S3 the latter tentatively suggests that indeed an exponent of approximately 1.2 ± 0.1 best fits the data.

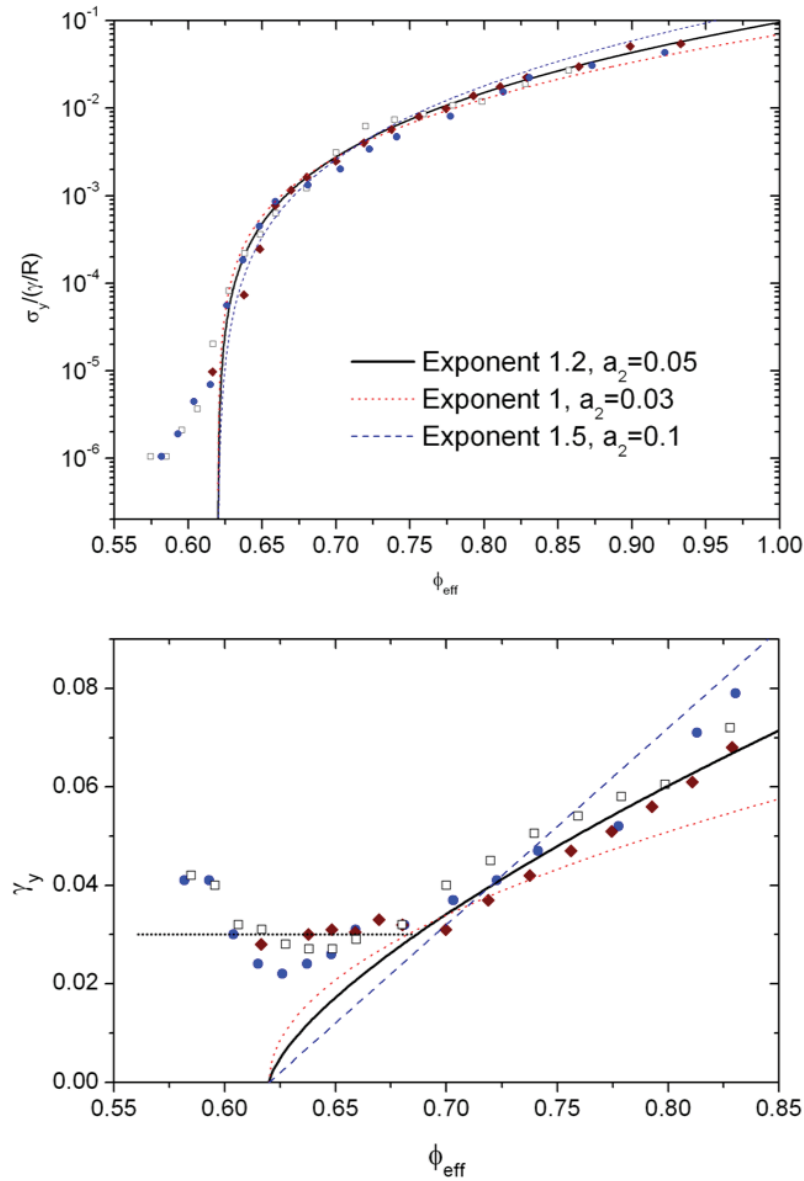


Figure S3 Yield stress (upper panel) and yield strain (lower panel) as a function of droplet volume fraction reproduced from Figures 1 and 3. The lines show the predictions using different exponents in Eq. (5) for the scaling of the yield stress via jamming ($\phi_J^{eff} = 0.62$). In order to obtain a fit to the data using Eq(1) we have allowed a_2 to vary.